Incommensurate Kekulé spiral order in magic-angle twisted bilayer graphene

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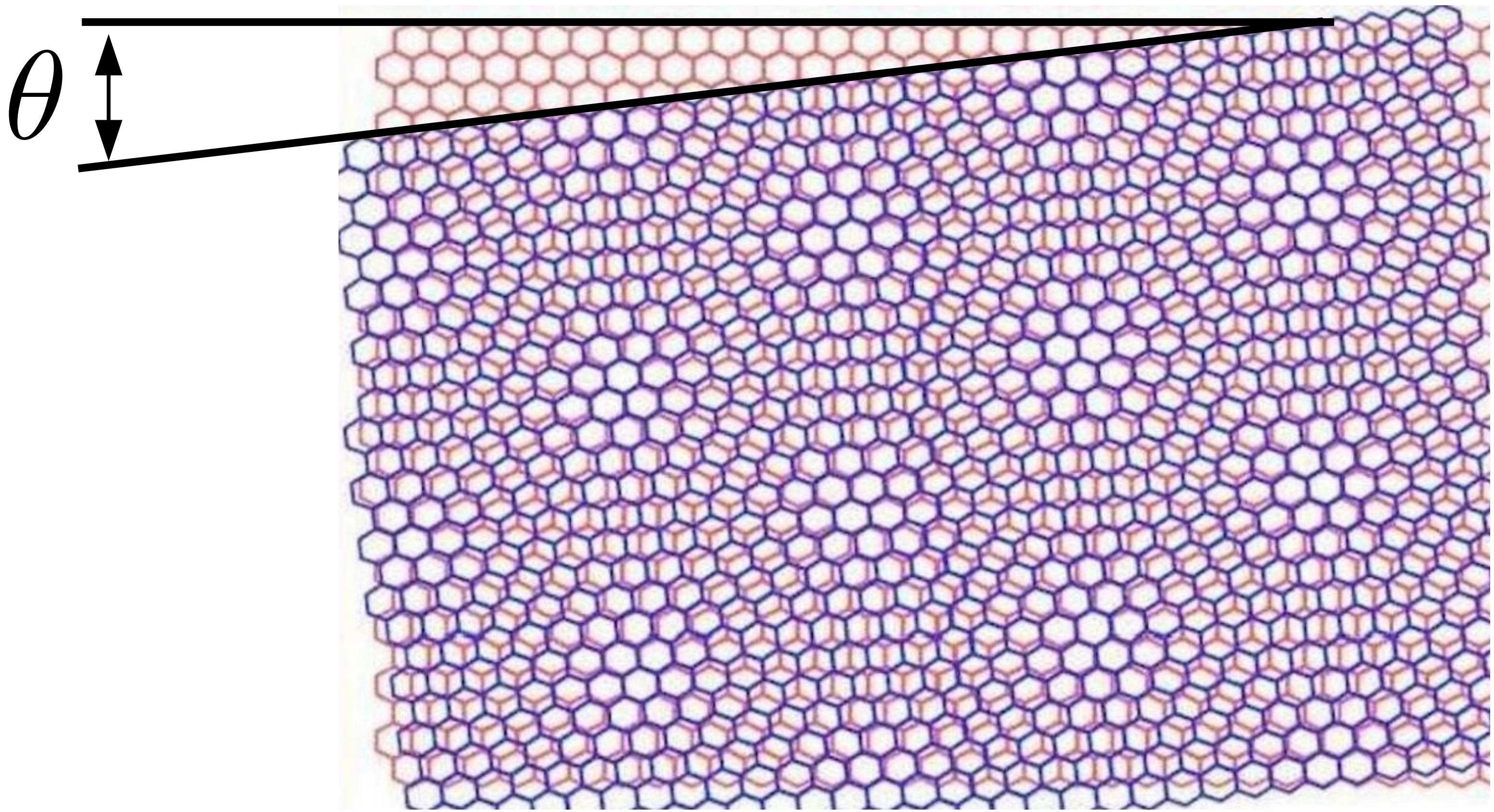
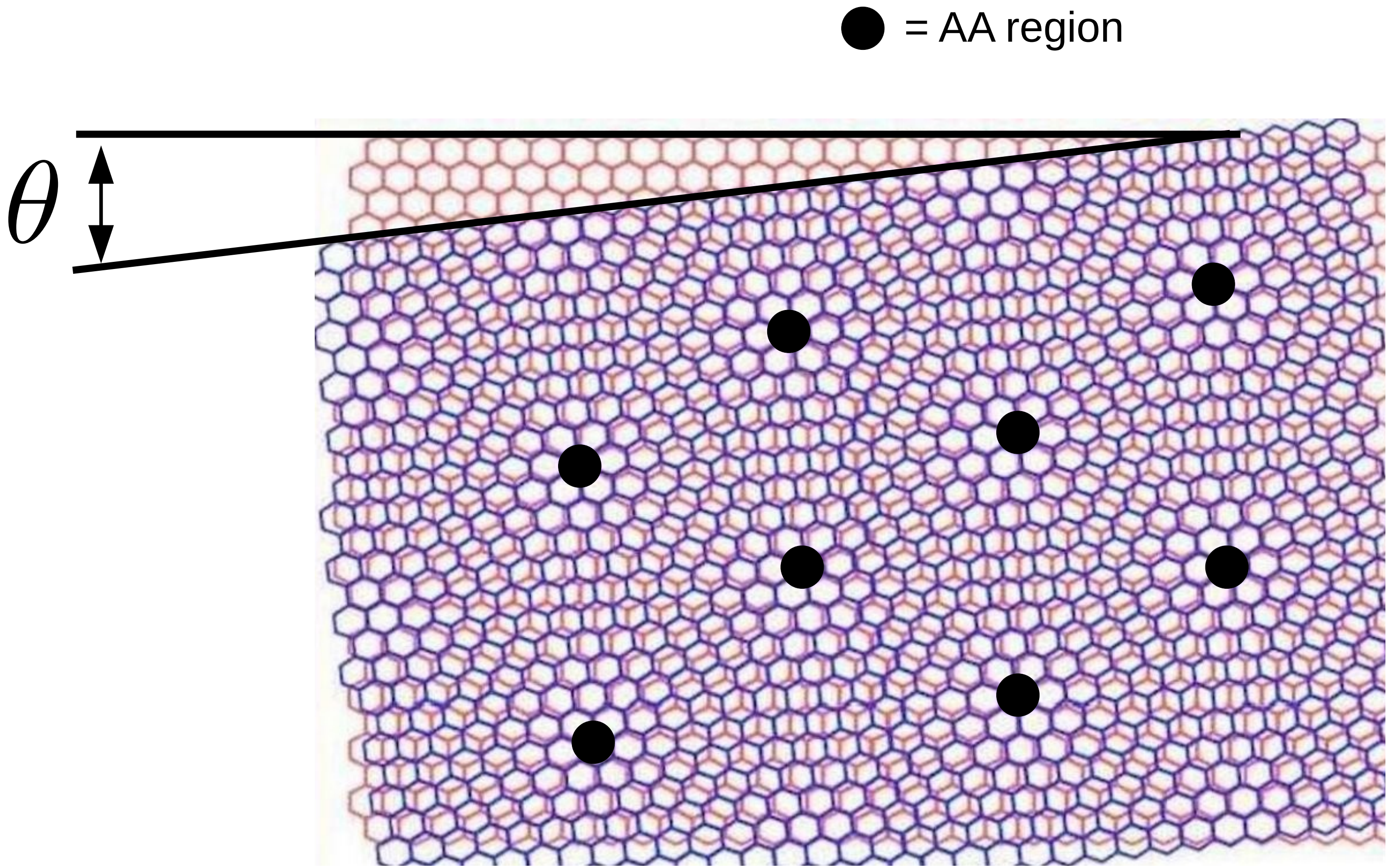
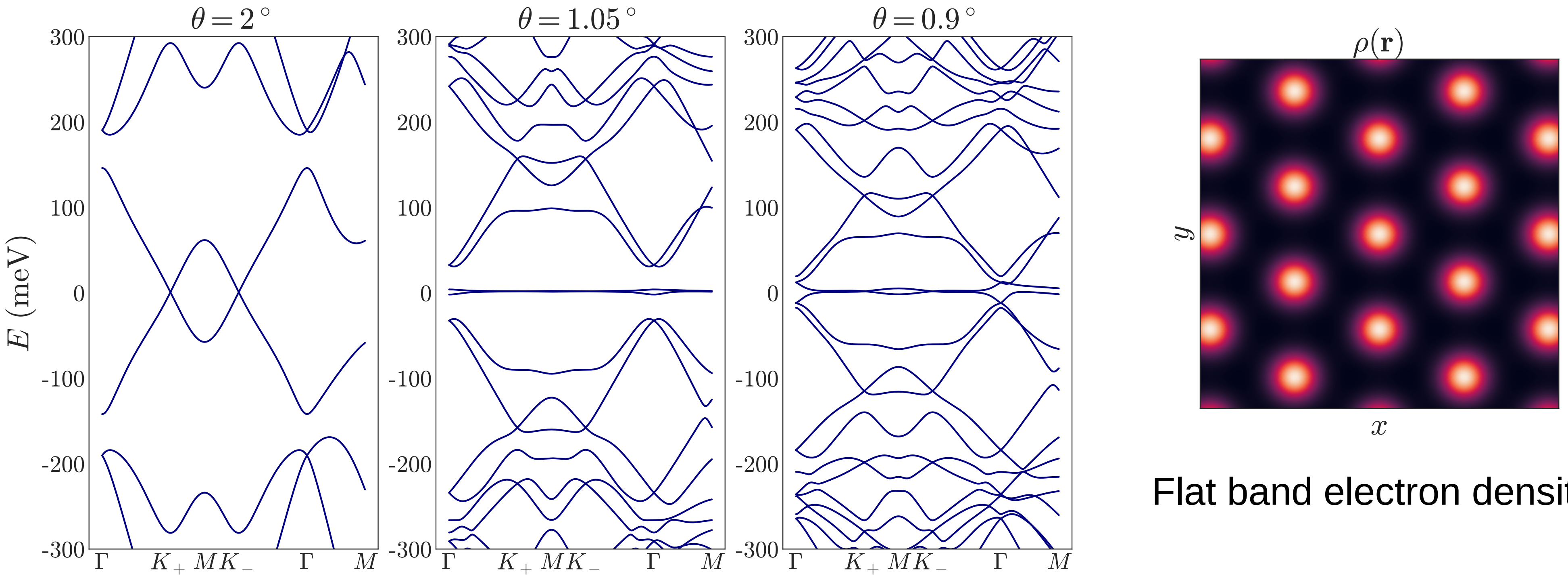
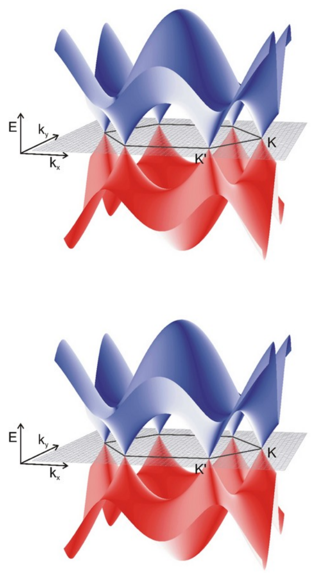

θ


= AA region
θ

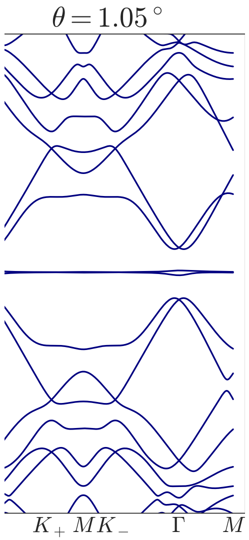


Flat band electron density

[Bistritzer, MacDonald (2011); dos Santos, Peres, Castro Neto (2012); Shallcross, Sharma, Kandelaki, Pankratov (2010)]

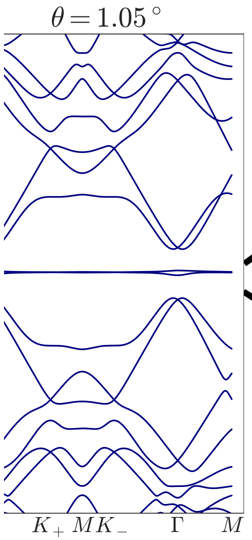


Mono-layer dispersion

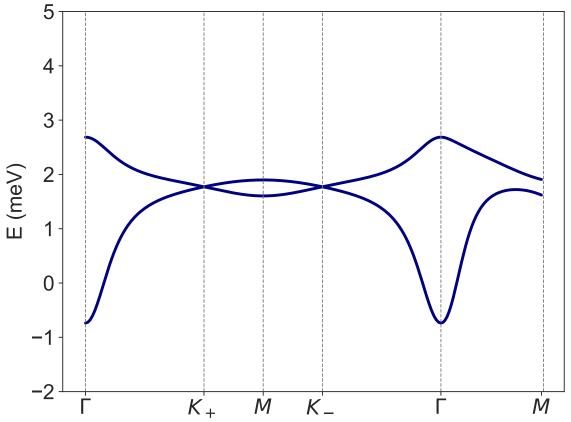


Valley K

TRS



Valley K'

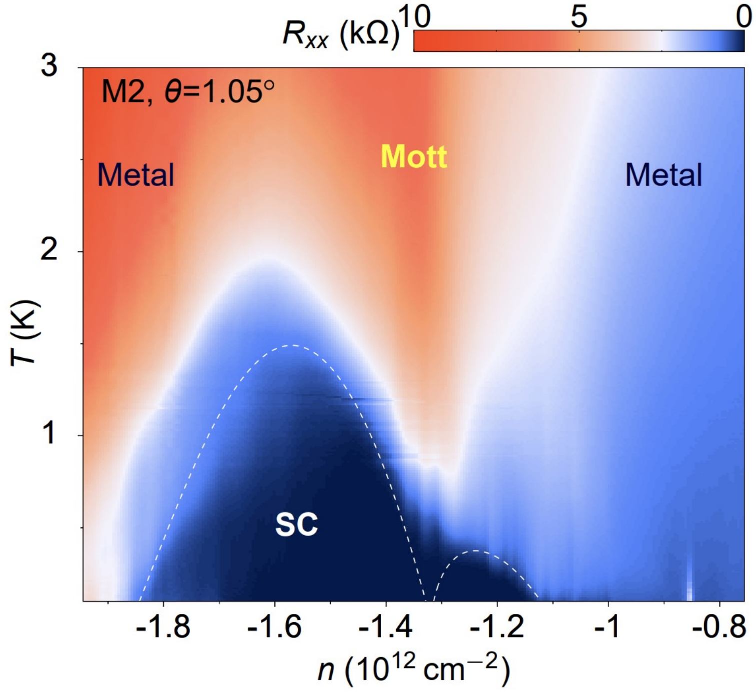


Band insulators at electron densities $\nu = \pm 4$

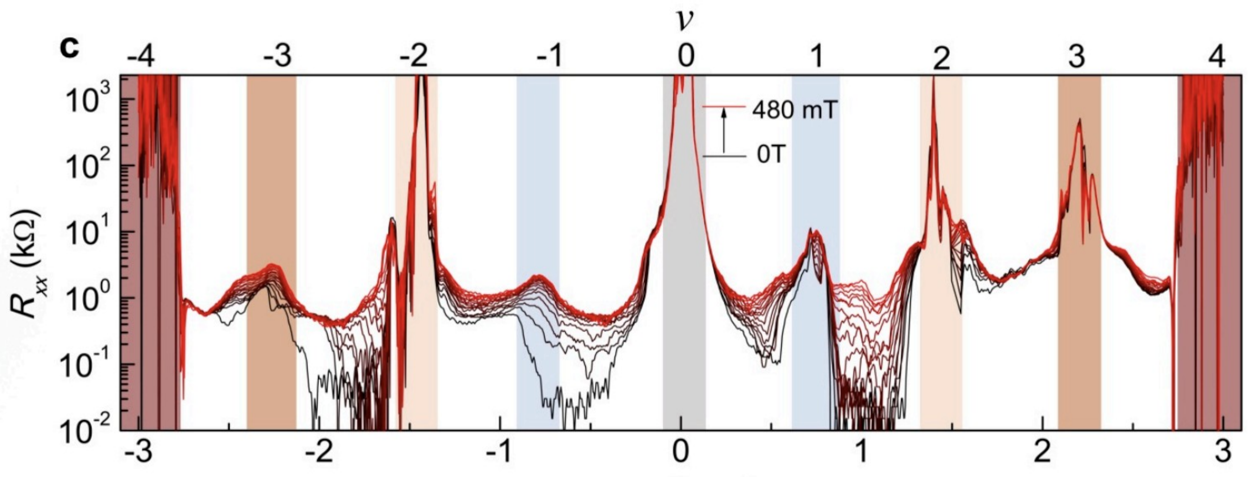
(4 electrons or holes per superlattice unit cell)

Valley U(1) symmetry
+ independent spin rotations in both valleys.

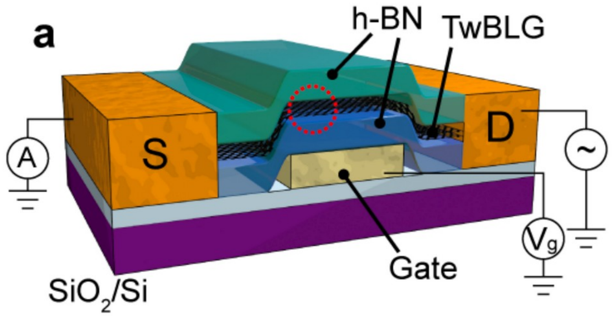
$\rightarrow$ Total (continuous) symmetry group is **U(2)xU(2)**.



Cao, Fatemi, …, Jarillo-Herrero *Nature* (2018)



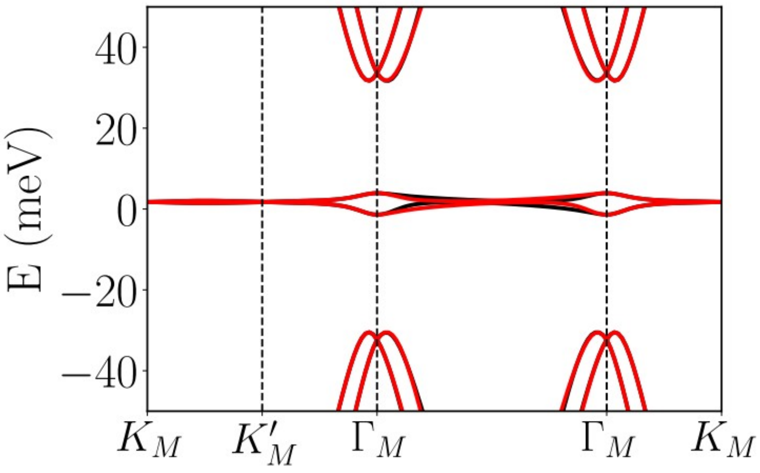
Lu, Stepanov, …, Efetov *Nature* (2019)



“Chiral” Limit of Bistritzer-MacDonald (BM) Model

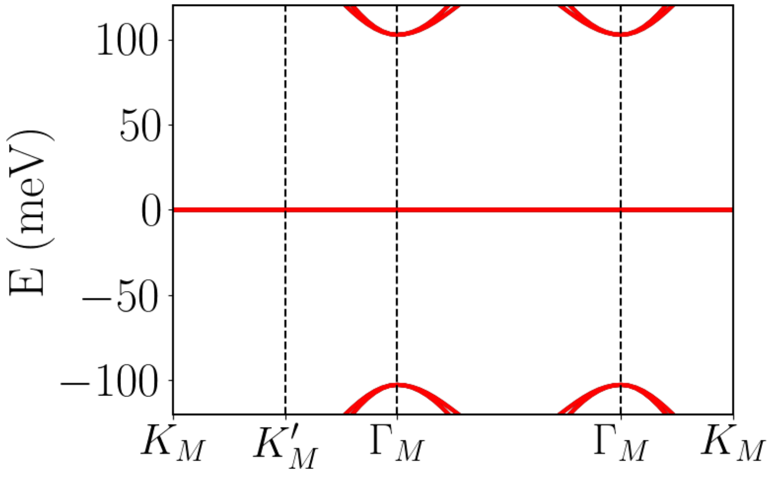
- **chiral ratio** $\eta = w_{AA}/w_{AB}$

$\eta = 0.8$



chiral limit

$\eta = 0$

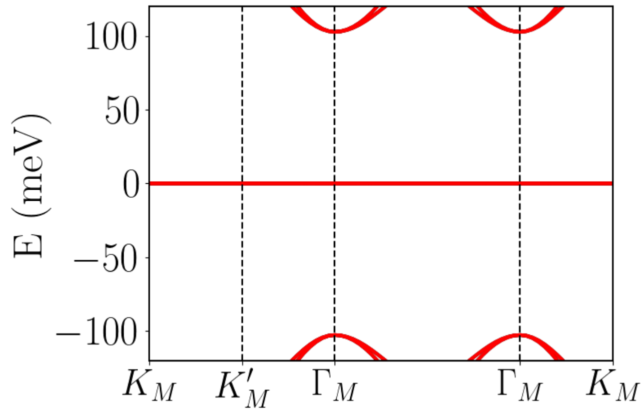


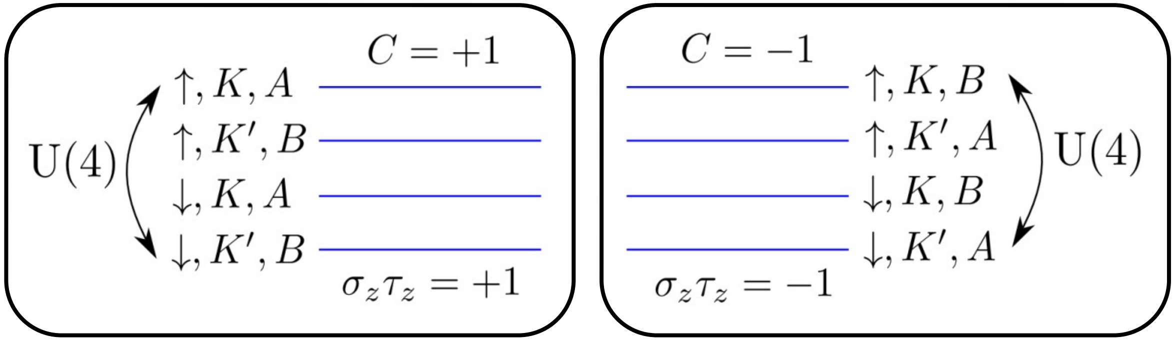
Tarnopolsky, Kruchkov, Vishwanath *PRL* (2019)

Chiral Limit (Strong Coupling)

- Bands labeled by sublattice, valley, spin $\{\sigma, \tau, s\}$
- Chern number $C = \tau_z \sigma_z$
- huge global symmetry $U(4)_{C=+1} \times U(4)_{C=-1}$

$\eta = 0$



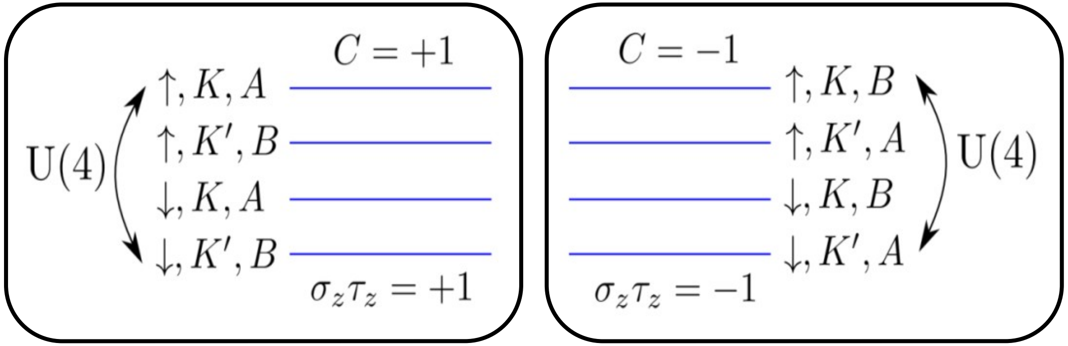


two ‘Chern quartets’
resembles multicomponent LL

$\Rightarrow$ Expect flavor ferromagnetism

NB, **Eslam Khalaf**, Liu, Chatterjee, Zaletel, Vishwanath *PRX* (2020)

Chiral Limit (Strong Coupling)



- add repulsive interactions
- ignore dispersion
- **Slater determinant ground states**

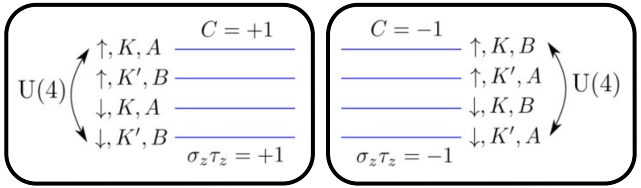
just fill any $4 + \nu$ out of 8 central Chern bands (generalized ferromagnets)

e.g. $|\Psi_{\mathrm{QAH}}\rangle = |KA\uparrow\rangle \otimes |KA\downarrow\rangle \otimes |K'B\uparrow\rangle \otimes |K'B\downarrow\rangle$

NB, **Eslam Khalaf**, Liu, Chatterjee, Zaletel, Vishwanath *PRX* (2020)

See also: Kang Vafek *PRL* (2019), Bernevig et al., *PRB* (2021); Lian et al., *PRB* (2021)

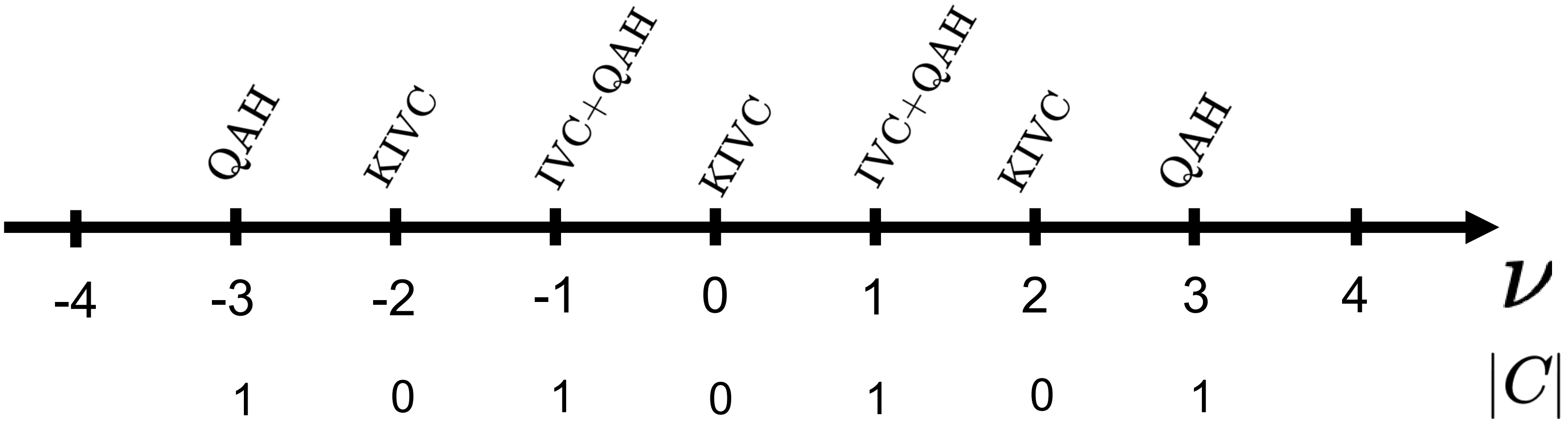
Chiral Limit (Strong Coupling)



- <u>single-particle terms</u> and <u>deviations from chiral limit</u> treated perturbatively
  $\rightarrow$ anisotropies in $U(4) \times U(4)$ manifold

select out subset of generalized ferromagnets
i.e. **'strong-coupling' insulators**

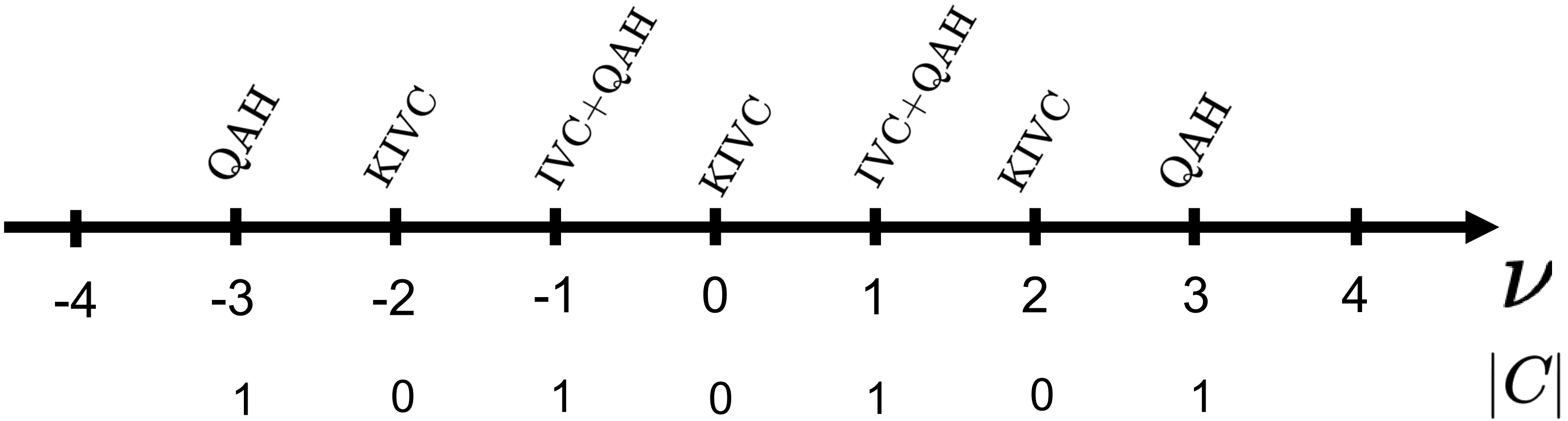
QAH = Quantum Anomalous Hall
IVC = Intervalley Coherent
KIVC = Kramers IVC



NB, Khalaf et al., *PRX* '20
Xie & MacDonald, *PRL* '20
Cea & Guinea, *PRB* '20
Zhang et al., *PRB '20*
Lian et al., *PRB* '21
Xie et al., *PRB* '21
Potasz et al., *PRL* '21
Parker et al., *PRL* '21
Kwan et al., *PRX* '21
…

Chiral limit strong-coupling & Experiments

- ✓ correlated insulators at integer ν as flavour ferromagnetism
- ✗ **experimentally CNP is often semimetallic**, but large-gap robust insulators in strong-coupling
- ✗ experimentally metallic/weak anomalies at $\nu = \pm 1$, but gapped insulators in strong-coupling
- ✗ observation of C=0 insulators at $\nu = 3$ [Pierce et al., Nat Phys, '21]



Beyond Chiral limit Strong Coupling

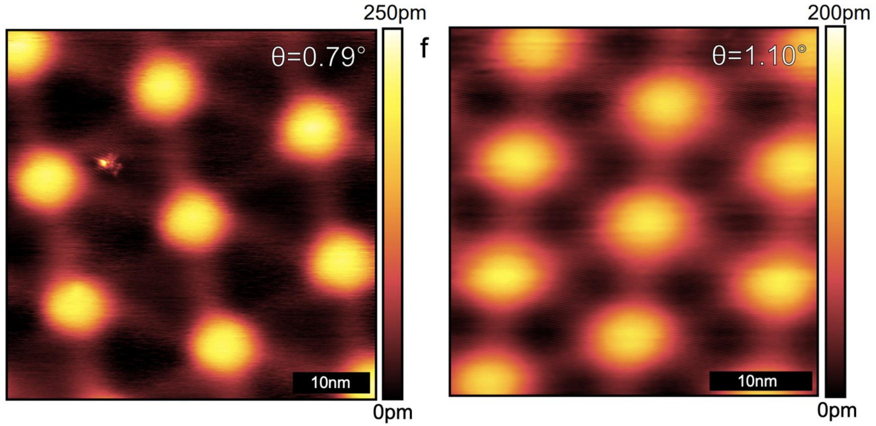
"Perturbations"

- Nonzero chiral ratio
- p-h sym breaking terms
- substrate potential
- Strain

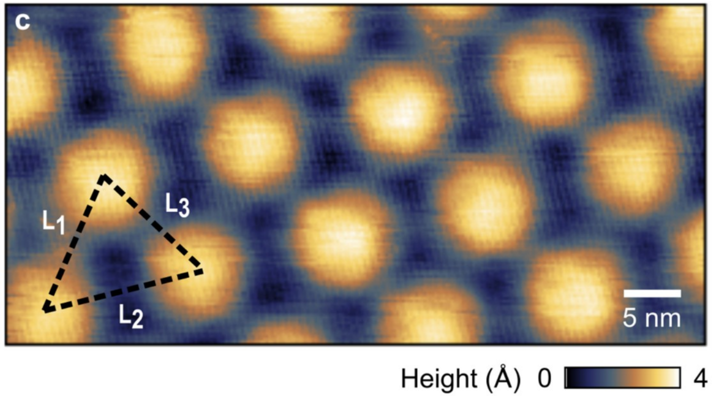
Treat these more seriously from the start?

The importance of strain

Kerelsky, McGilly, …. , Dean, Rubio, Pasupathy *Nature* (2019)



Xie, Lian, … , Bernevig, Yazdani *Nature* (2019)



Heterostrain of magnitude $\epsilon \sim 0.1 - 0.7\%$ observed in STM.

This is small, but moiré patterns act like a magnifying glass for strain.

To lowest order, strain couples to mono-layer graphene as a vector potential.

$$h_D(\mathbf{k}) = \hbar v_F (M[\mathbf{k} - \mathbf{A}]) \cdot \boldsymbol{\sigma}$$

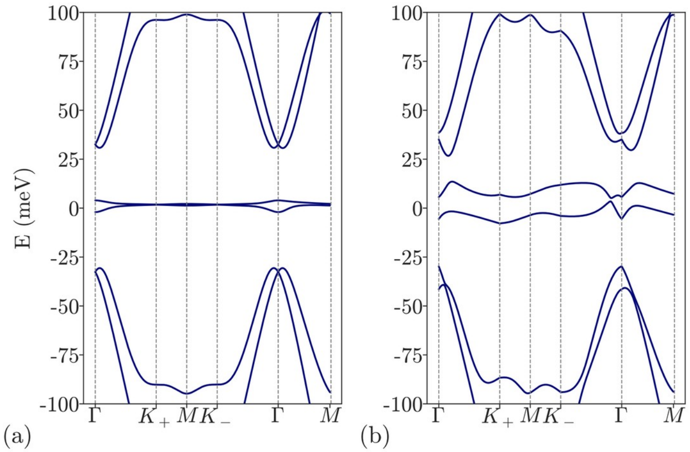
$$M = \begin{pmatrix} 1+\epsilon_{xx} & \epsilon_{xy} \\ \epsilon_{xy} & 1+\epsilon_{yy} \end{pmatrix}$$

Suzuura, Ando *PRB* (2002); Sasaki, Saito (2008)

$$\boldsymbol{A} = \frac{\beta}{2a}(\epsilon_{xx} - \epsilon_{yy}, -2\epsilon_{xy})$$

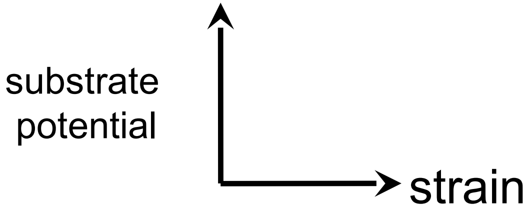
Effect of strain on MATBG band spectrum (DPs in a single valley are no longer related by symmetry):

Without strain →

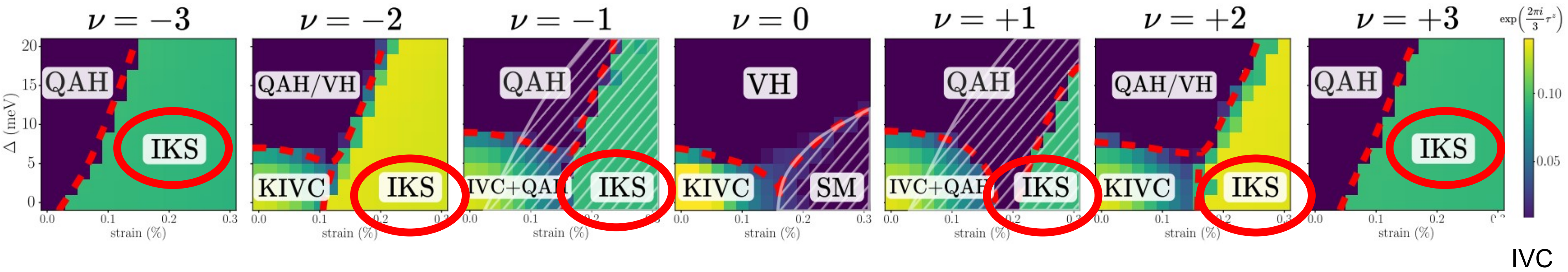


← With the experimentally observed strain

Bi, Yuan, Fu *PRB* (2019)



<u>IKS: Incommensurate Kekulé Spiral</u>
QAH: quantized anomalous Hall state
KIVC: Kramers intervalley coherent state
VH: valley Hall state
IVC: intervalley coherence
SM: semimetal
Hatched: no HF gap

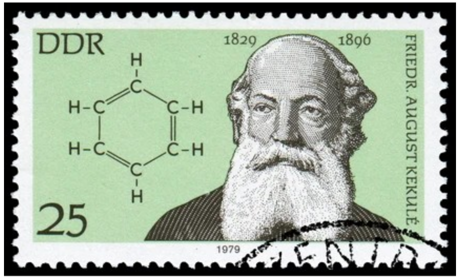


- self-consistent Hartree-Fock with realistic parameters
- check for completely general translational symmetry-breaking
- **A new type of order (IKS order) dominates all non-zero integers with strain**

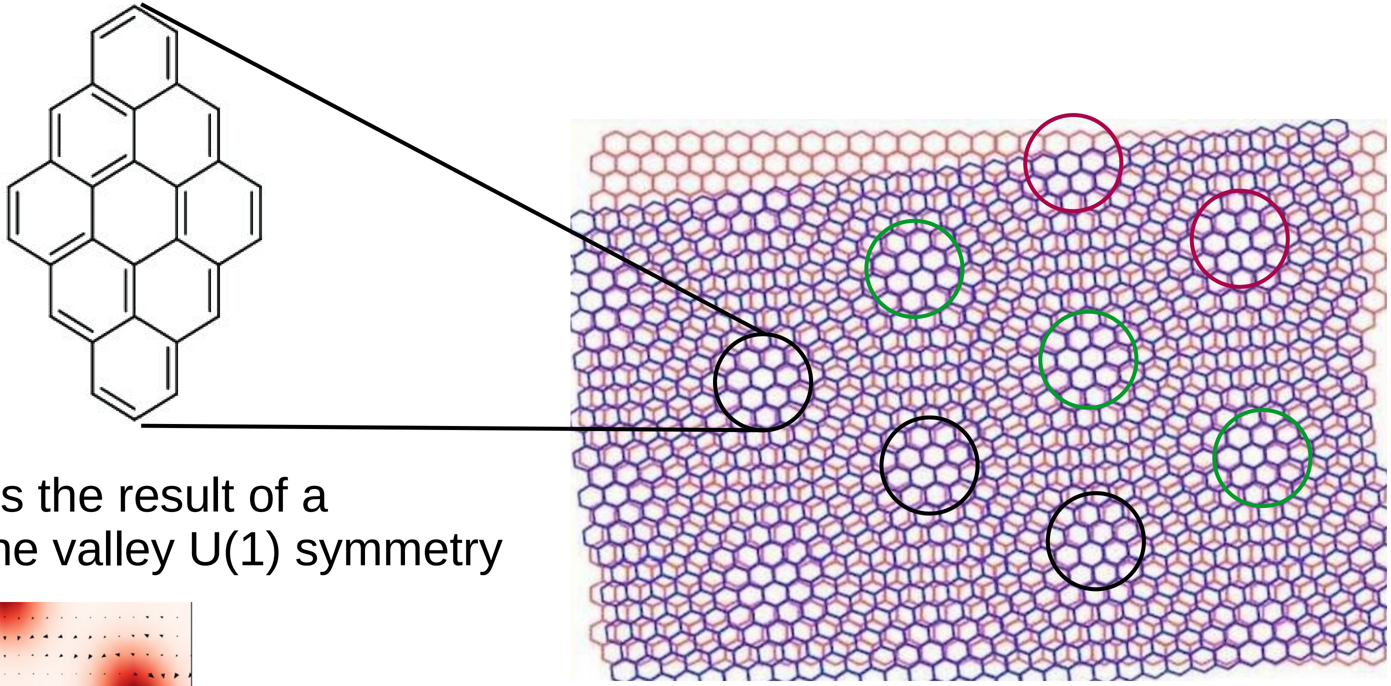
Kwan, Wagner, Simon, Parameswaran, NB *PRX* (2021)

What is IKS?

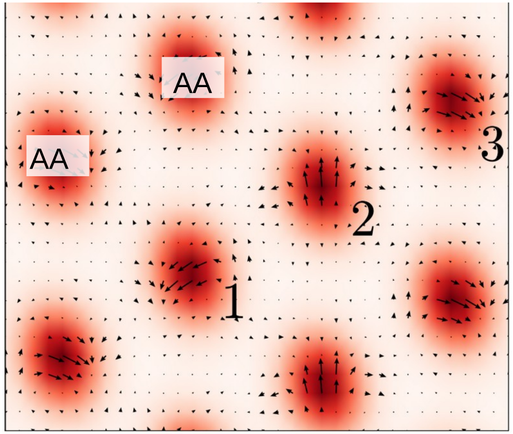
Kekulé pattern:



shutterstock.com · 438531103



$\sqrt{3} \times \sqrt{3}$ Kekulé pattern is the result of a spontaneous breaking of the valley U(1) symmetry

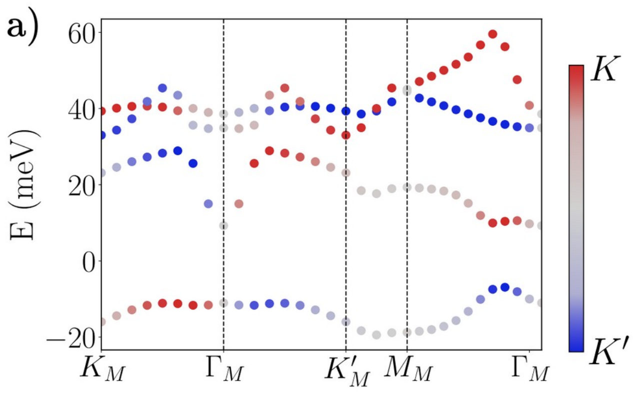


The Kekulé pattern modulates on the superlattice scale with an incommensurate wavevector

The IKS order has a non-zero wavevector and thus breaks translation symmetry. However, it preserves a modified translation symmetry:

$$\hat{T}'_{\mathbf{a}_i} = \hat{T}_{\mathbf{a}_i} e^{i\mathbf{a}_i \cdot \mathbf{q}\tau^z}$$

Mean-field IKS band structure at $\nu = -2$



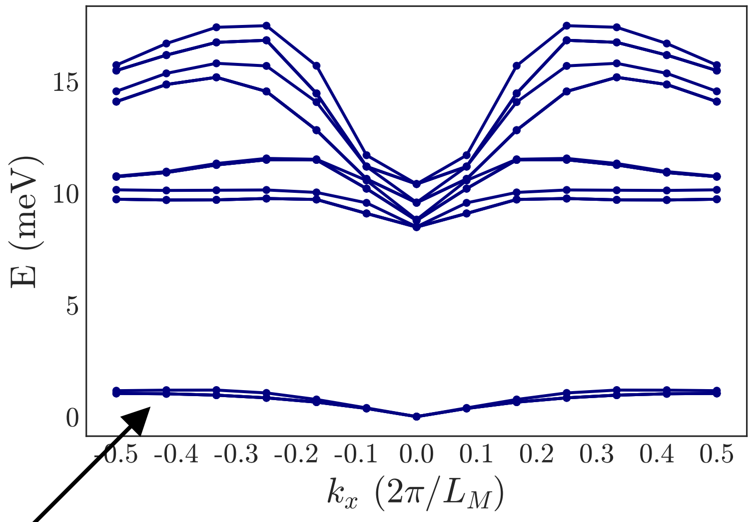
This implies a generalized Bloch theorem:

$$\psi_{\tilde{\mathbf{k}}}(\mathbf{r}) = e^{i\mathbf{r}\cdot(\tilde{\mathbf{k}} - \mathbf{q}\tau^z/2)} u_{\tilde{\mathbf{k}}}(\mathbf{r})$$

$$u_{\tilde{\mathbf{k}}}(\mathbf{r} + \mathbf{a}_i) = u_{\tilde{\mathbf{k}}}(\mathbf{r})$$

Beyond mean-field theory, the modified translation symmetry pins IKS insulators to integer superlattice fillings as the result of a generalized Lieb-Schultz-Mattis theorem

Collective modes of the IKS at $\nu = -2$: (This state has zero spin polarization.)

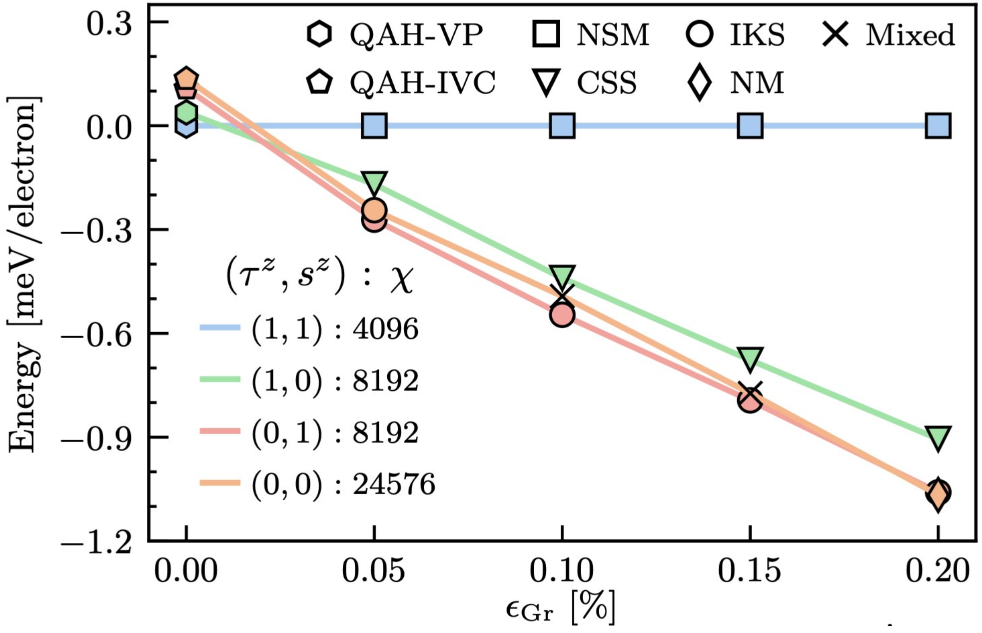


4 Goldstone modes! (one singlet, one triplet)

Broken symmetry generators: $\tau^z, s^x\tau^z, s^y\tau^z, s^z\tau^z$

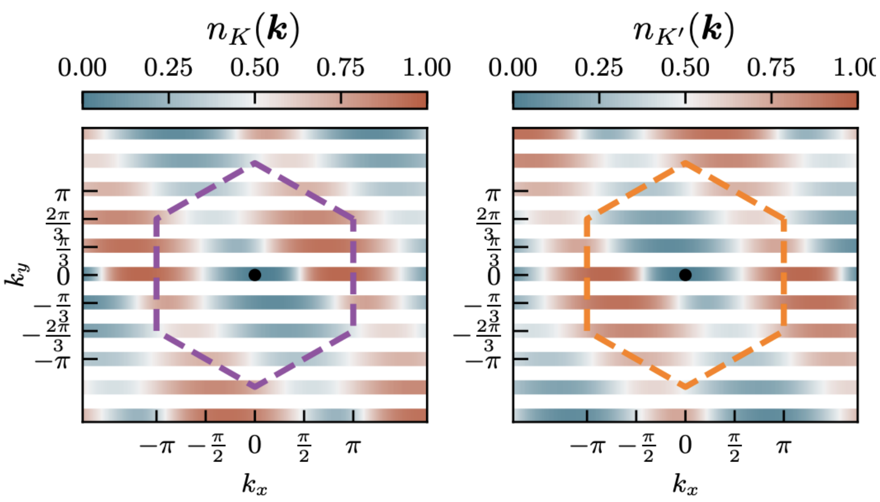
IVC states, spin polarized or unpolarized, always have Goldstone modes associated with spin fluctuations.

DMRG results on strained interacting BM model at $\nu = -3$:



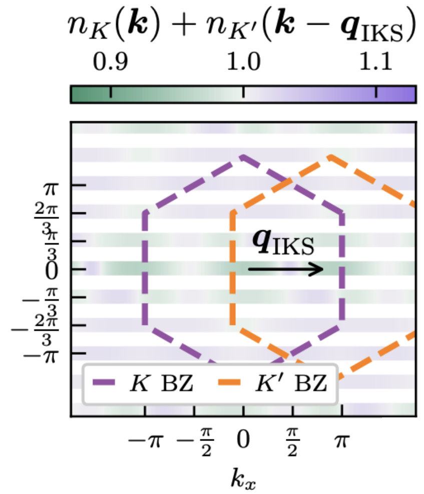
Strain

Wang, Parker, Soejima, Hauschild, Anand, NB, Zaletel (2023)

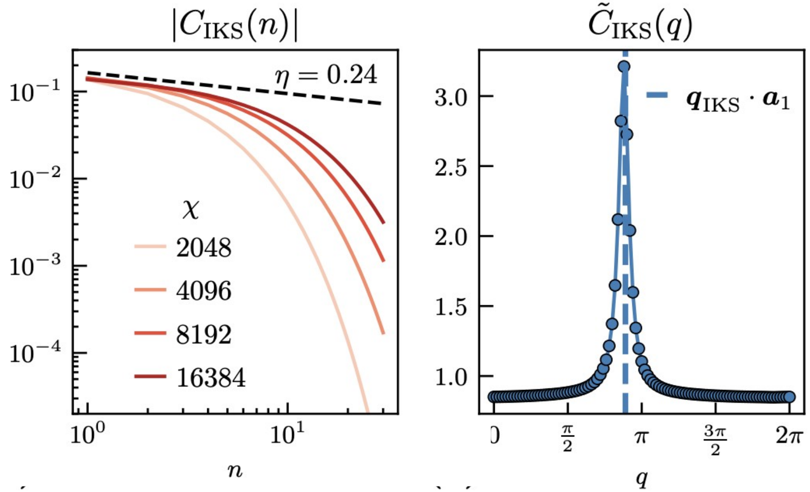


Valley-resolved occupation numbers

Shift and superimpose



Correlation function of IKS order parameter



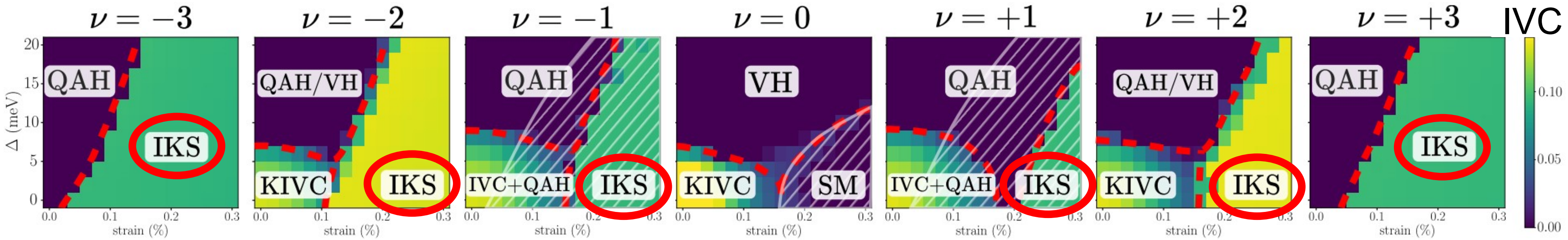
Wang, Parker, Soejima, Hauschild, Anand, NB, Zaletel (2023)

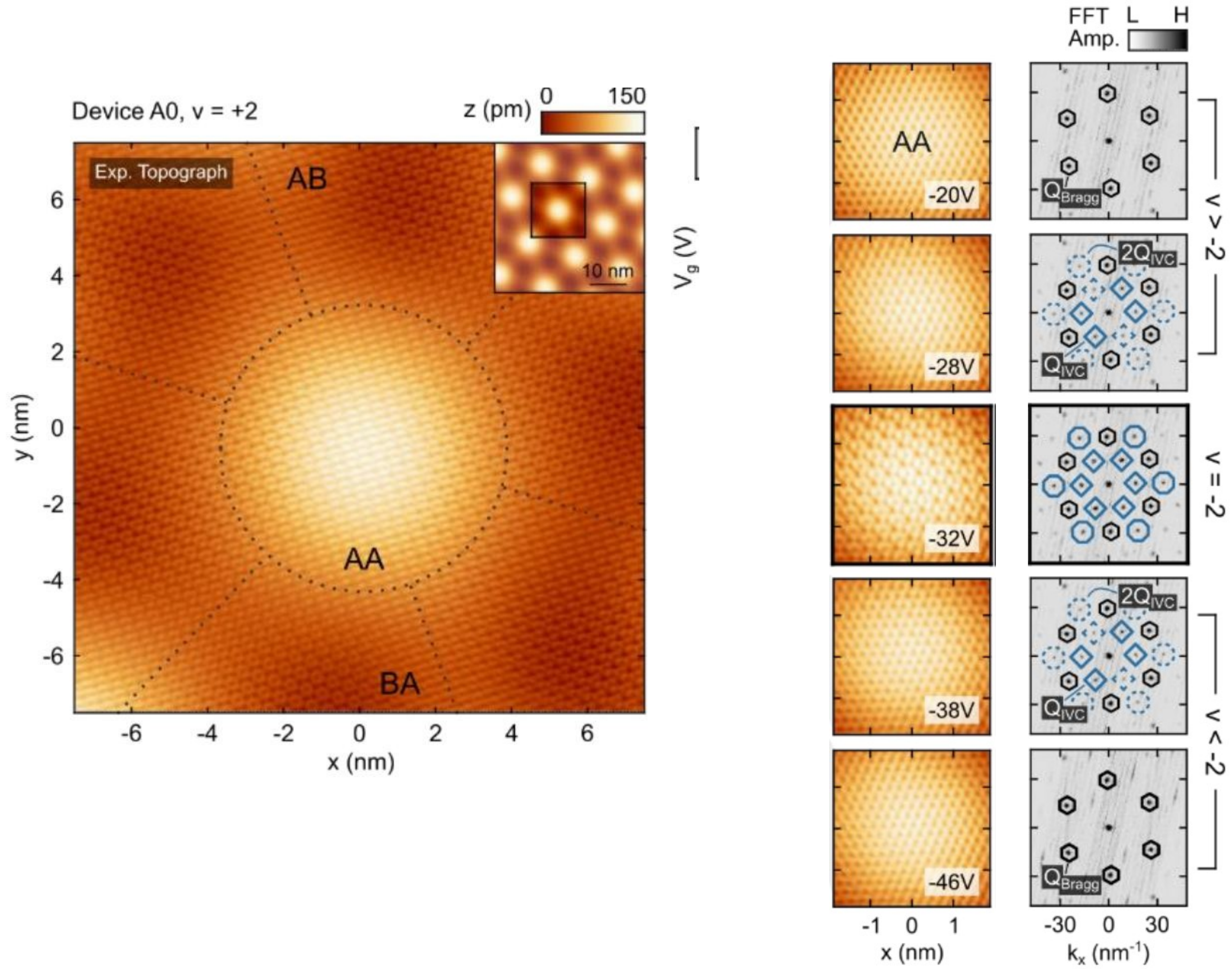
Connections of IKS to experiment

- ✓ strain significantly degrades gap and induces semimetallic behaviour at CNP
- ✓ IKS at $\nu = \pm 1$ is a ‘near-insulator’
- ✓ gapped IKS at $\nu = \pm 2$ is very robust
- ✓ gapped IKS at $\nu = \pm 3$ has C=0 Pierce et al., Nat Phys, ’21
- ✓ IKS emerges at strain ratios well within experimental limits

✓ NEW DEVELOPMENT: KEKULÉ SPIRAL OBSERVED IN STM!

arXiv:2303.00024, Nuckolls et al (Yazdani group)





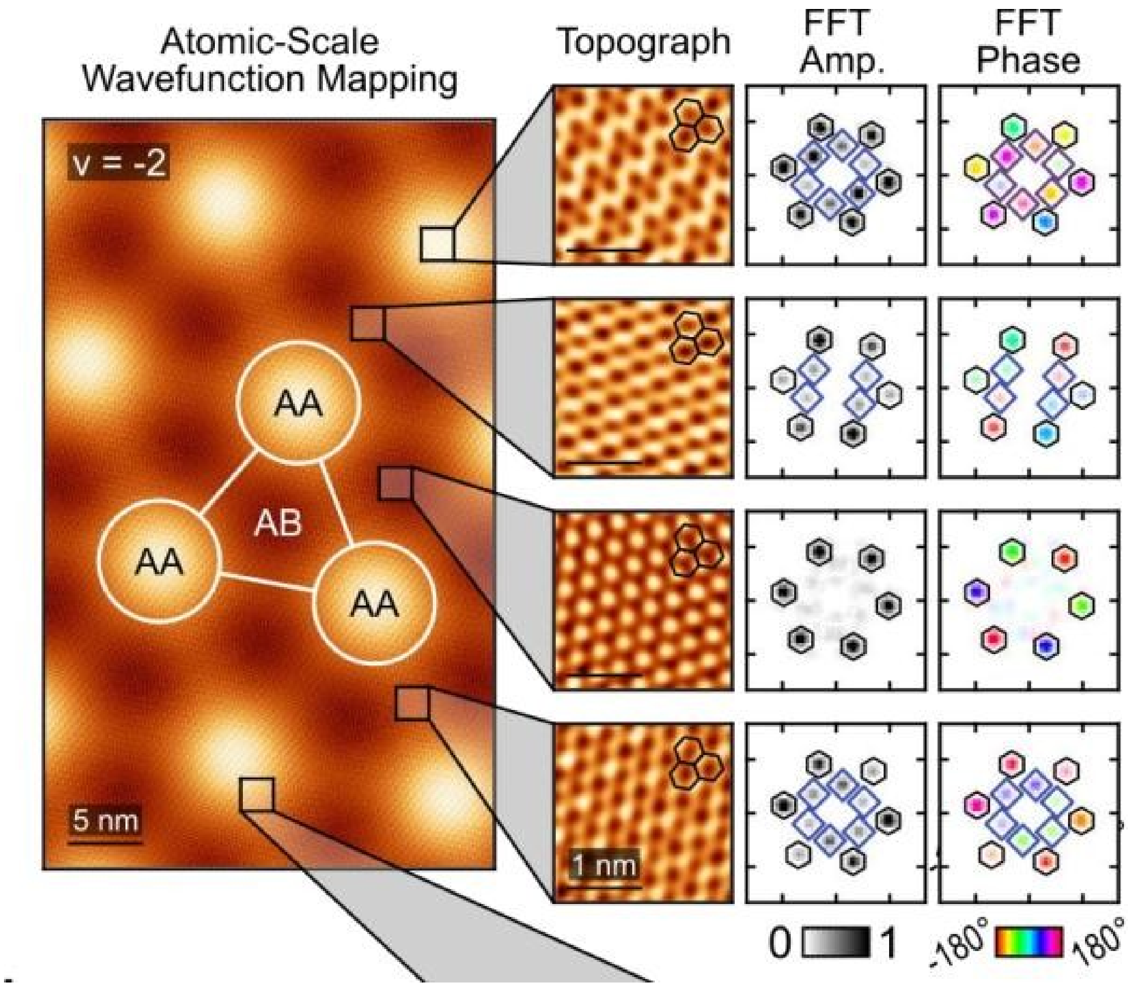
Graphene
Bragg
Peaks

Also Kekulé distortion

Kekulé distortion =
Intervalley Coherence,
.....BUT NOT KIVC!

Hong et al *PRL* 22
Calugaru et al *PRL* 22

arXiv:2303.00024, Nuckolls et al (Yazdani group)

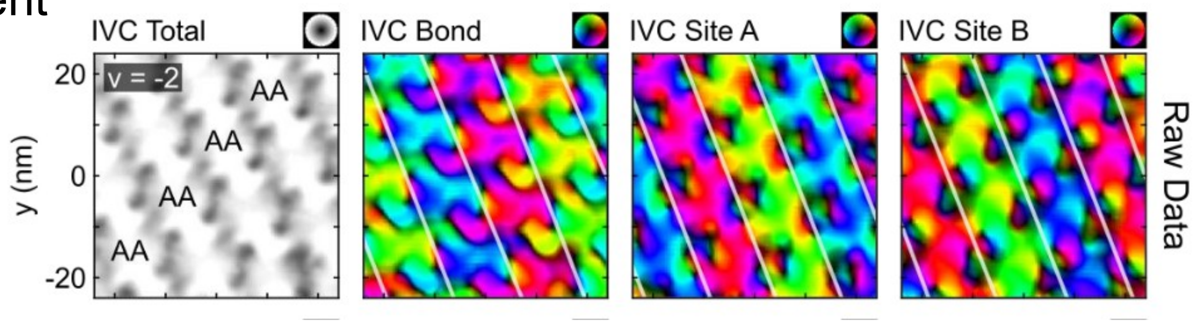


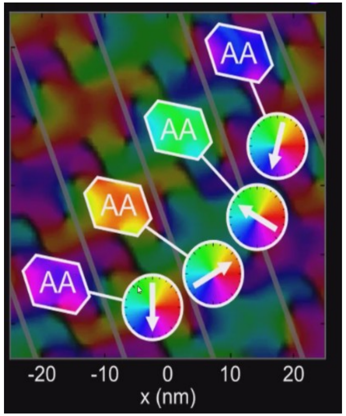
Kekule Bragg Peaks seen almost everywhere, but pattern changes

Analysis by phases of FFT peaks

arXiv:2303.00024, Nuckolls et al (Yazdani group)

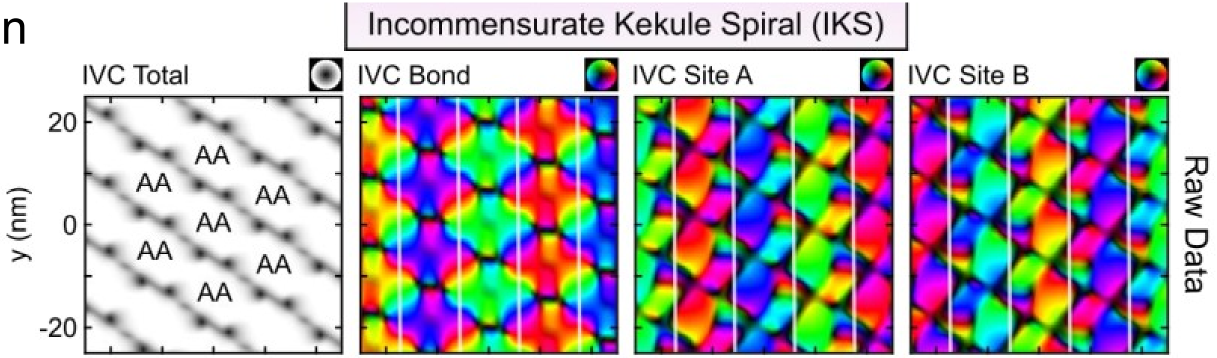
Experiment





(Screenshot from talk by Nuckolls/Yazdani)

Simulation



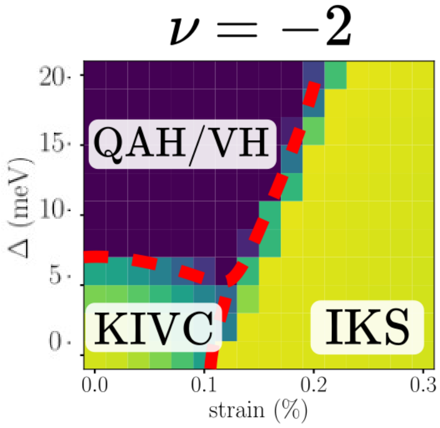
IKS is seen in all "typical" samples at n = ± 2.

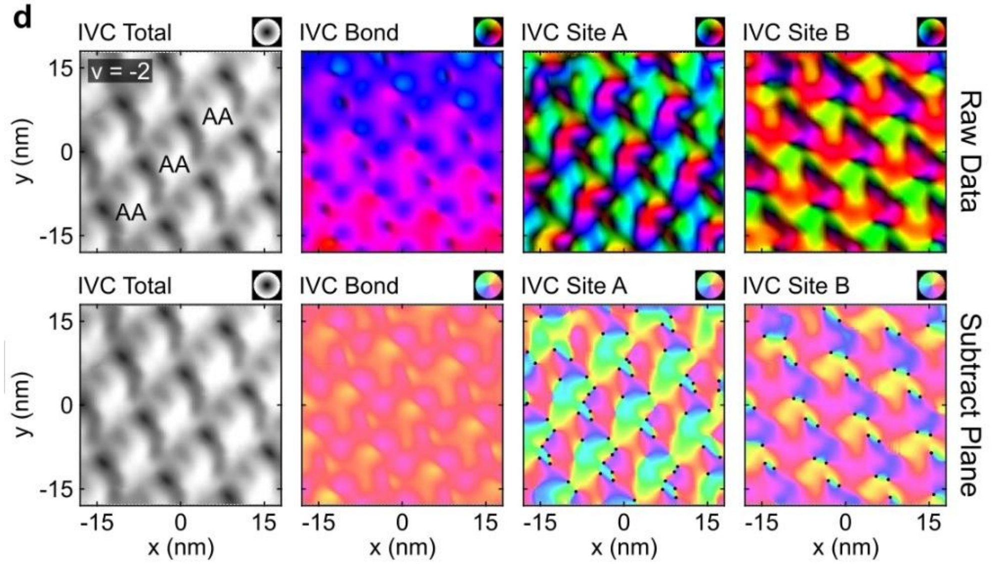
In a few samples with ultra-low strain (< 0.1%) they see something different (but not KIVC!)

arXiv:2303.00024, Nuckolls et al (Yazdani group)

What about highly unstrained samples?

Hartree-Fock says KIVC
--- should show no Kekulé density pattern





arXiv:2303.00024, Nuckolls et al (Yazdani)

...but data sees Kekulé density pattern (q=0)

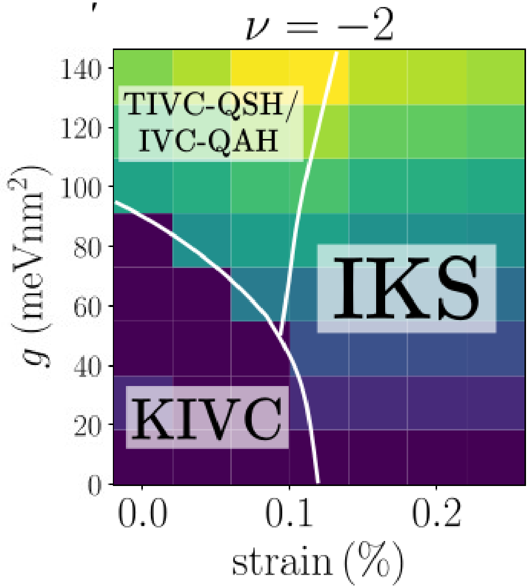
Electron-phonon coupling and competing Kekulé orders in twisted bilayer graphene

Yves H. Kwan,[1] Glenn Wagner,[2] Nick Bultinck,[3,4] Steven H. Simon,[3] Erez Berg,[5] and S.A. Parameswaran[3]

$$\hat{H} = \epsilon a^\dagger a + \gamma a + \gamma^* a^\dagger \longrightarrow \hat{H} = \epsilon b^\dagger b - \frac{|\gamma|^2}{\epsilon}$$

$$b^\dagger = a^\dagger + \gamma/\epsilon$$

An electron state with density modulation lowers total phonon energy.



TIVC-QSH $\big(|KA\uparrow\rangle + |\bar{K}B\uparrow\rangle\big)\big(|KB\downarrow\rangle + |\bar{K}A\downarrow\rangle\big)$

IVC-QAH $\big(|KA\uparrow\rangle + |\bar{K}B\uparrow\rangle\big)\big(|KA\downarrow\rangle + |\bar{K}B\downarrow\rangle\big)$

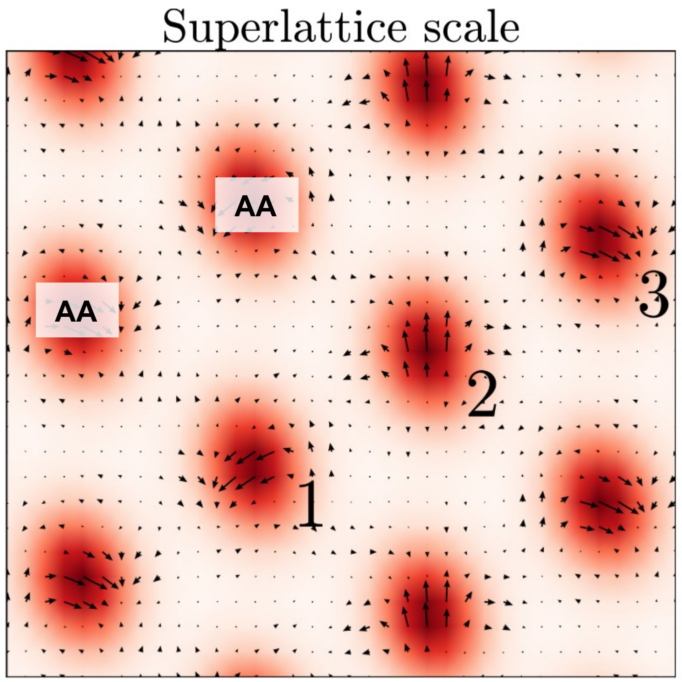
Summary:

IKS: Incommensurate **K**ekulé **S**piral

It is there in “typical” samples

Explains a lot.... how much more will it explain?

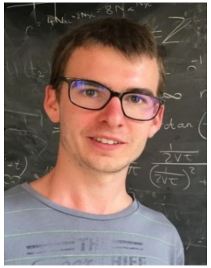
(teaser: IKS in TRI-layer twisted graphene reported by
Nadj-Perge group at Aspen.)



Thank you for listening!

Yves Kwan

Glenn Wagner



Steve Simon

Sid Parameswaran

Tomohiro Soejima

Michael Zaletel

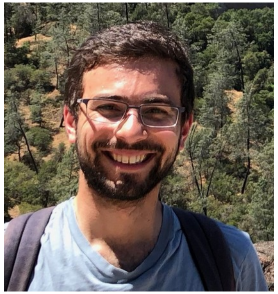
→ Princeton → Zurich Oxford Berkeley

DMRG team:

Tianle Wang

Sajant Anand

Daniel Parker



Johannes Hauschild

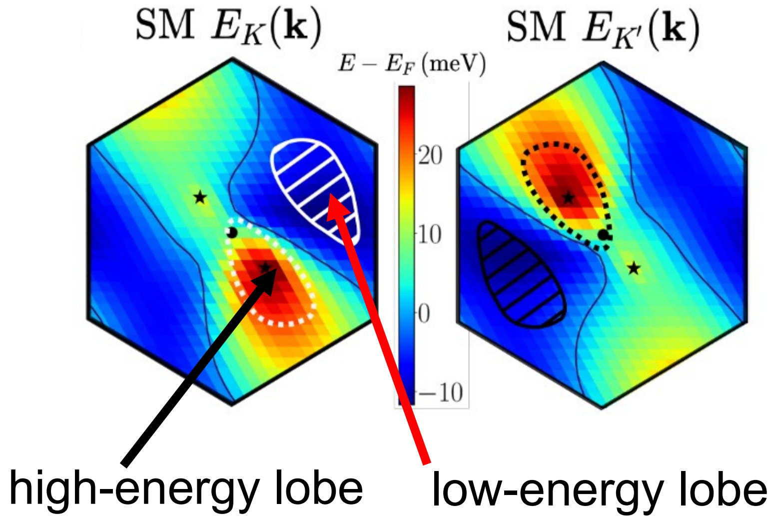
Berkeley Harvard TU Munich

Physical mechanism for IKS

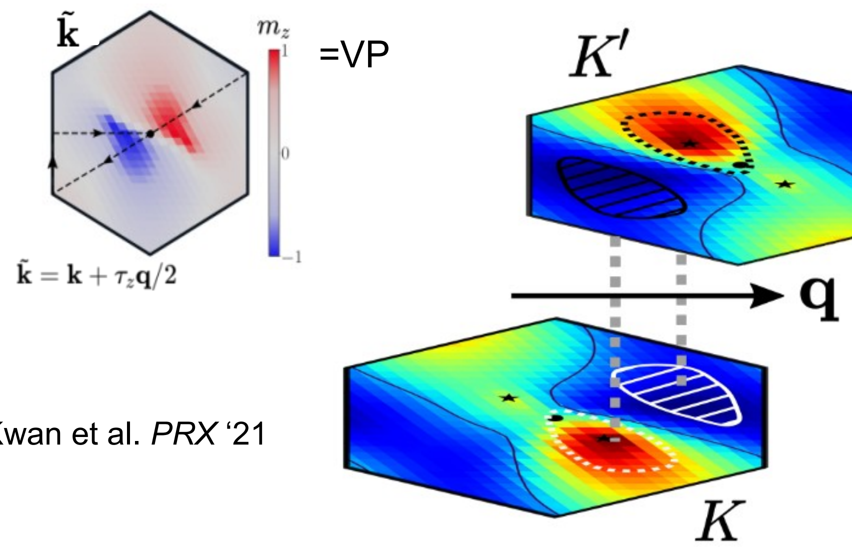
- **Interaction-induced band renormalization** important in TBG

Rademaker et al., *PRB* '19 Xie et al., *Nature* '19
Cea et al., *PRB* '19 Wong et al., *Nature* '20
Kang et al., *PRL* 21

consider HF bands of symmetric metal at $\nu = -2$:



- Align high-energy lobe in one valley with low-energy lobe in other valley to find **q**. IVC everywhere else.



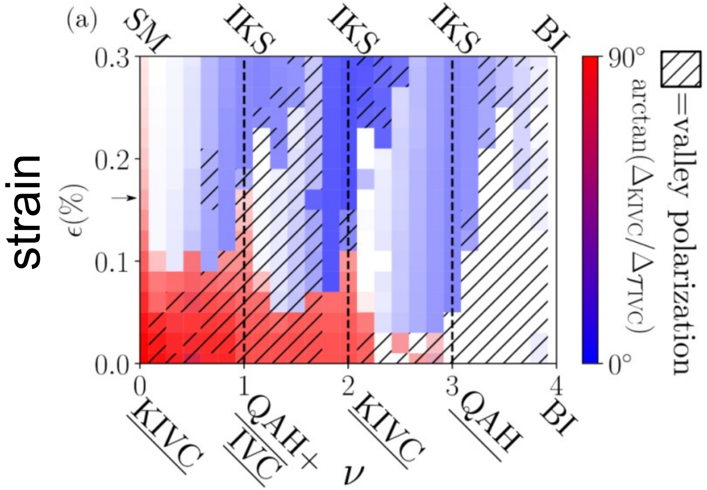
Kwan et al. *PRX* '21

Strain increases dispersion → Strong Coupling Ferromagnetism Fails!

Non-integer fillings

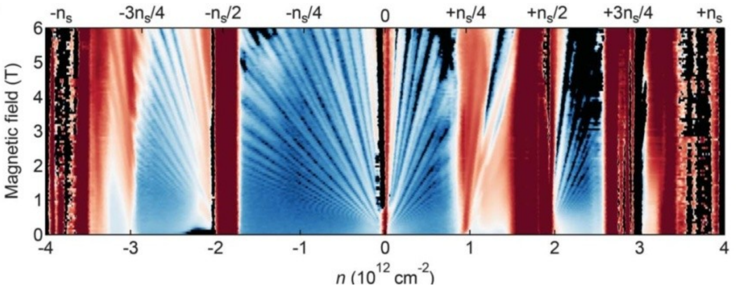
- IKS persists for a large range of dopings at finite strain

Wagner, Kwan, NB, Simon, Parameswaran *PRL* (2022)

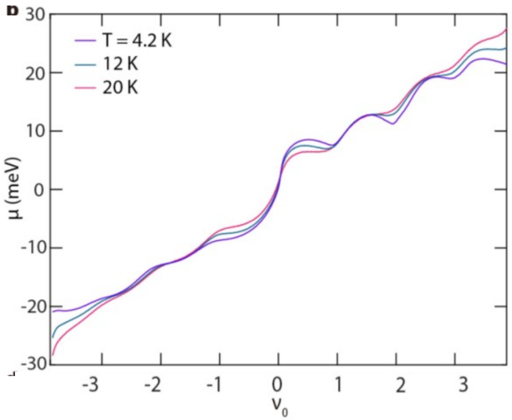


✔ Landau fan degeneracy at finite strain is consistent with experiments
4,-,2,1 degeneracy at fillings 0,1,2,3

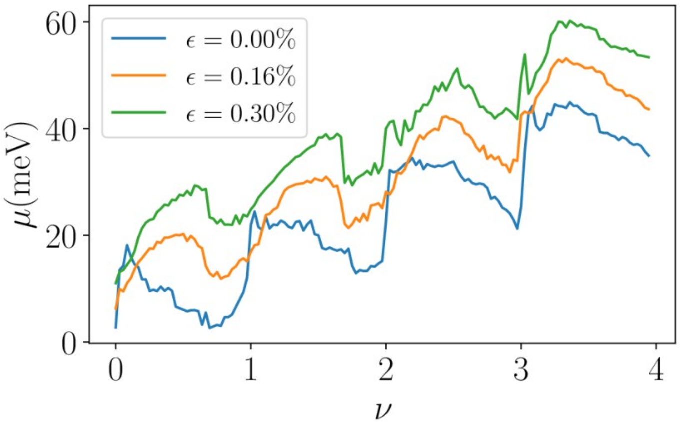
✔ ‘cascade’ transitions consistent with experiment [Zondiner et al., *Nature* ‘20]



Yankowitz et al., *Science* ‘19

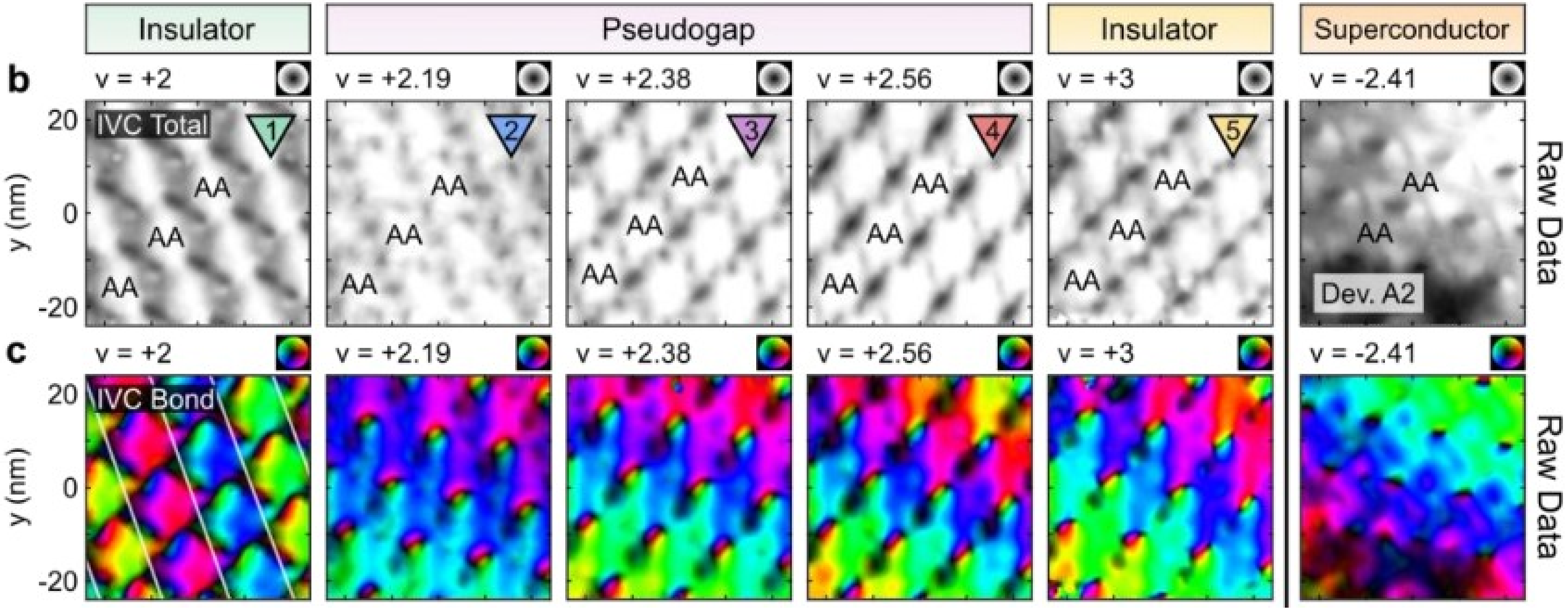


Saito et al., *Nature* ‘21



Wagner et al. *PRL* (2022)

Non-integer fillings



IKS-ish?

arXiv:2303.00024, Nuckolls et al (Yazdani group)